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METALLURGICAL PROCESSES IN MULTI-COMPONENT RARE
EARTH-TRANSITION METAL PERMANENT MAGNET ALLOYS(U)
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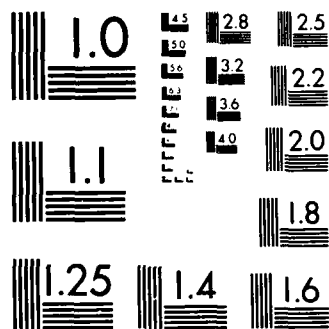
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METALLURGICAL PROCESSES IN MULTI-COMPONENT
RARE EARTH-TRANSITION METAL PERMANENT MAGNET ALLOYS

FINAL REPORT

A. E. RAY

JUNE 1987

PREPARED FOR
U.S. ARMY RESEARCH OFFICE

CONTRACT NO. DAAG29-84-K-0012
21026-EL

PREPARED BY:

UNIVERSITY OF DAYTON
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PROBLEM STATEMENT

Permanent magnets which produce higher magnetic fields at high temperatures and higher and essentially temperature independent fields over wide temperature ranges than those presently available are required to implement improved designs for applications such as traveling wave tubes and inertial guidance devices. Two types of rare earth-transition metal permanent magnets are presently employed for applications requiring high and stable performance at elevated temperatures, those based on SmCo_5 (1:5 type) and those based $\text{Sm}_2(\text{Co,Fe})_{17}$ (2:17 type). In both cases, the negative temperature dependences of the magnetic properties are minimized by the partial substitution of a heavy rare earth, such as Gd, for the Sm. A third type of rare earth-transition permanent magnets, those based on $\text{Nd}_2\text{Fe}_{14}\text{B}$, are not employed for such applications due to the severe deterioration of their magnet properties at elevated temperatures.

Uncompensated 1:5 type permanent magnets have been commercially available since the early 1970's, and temperature compensated versions since the mid-1970's. The 1:5 magnets have been developed to where they are now considered mature products by the magnet industry and significant improvements in their performance is not anticipated. Uncompensated 2:17 type permanent magnets have been commercially available since about 1980, and temperature compensated 2:17's have gradually become available over the past several years. The temperature compensated 2:17's have begun to displace the temperature compensated 1:5's as the magnets of choice for some high performance applications [1]. In contrast to the 1:5's, significant improvements of the magnetic properties of the 2:17's seem possible, [2] even though the record energy product for a 2:17-type permanent magnet, $(\text{BH})_{\text{max}} = 33 \text{ MGOe}$, was announced in 1980 [3]. Indeed, the 2:17's may be the best, if not the only, source for substantial improvements in high temperature performance of permanent magnets in the near term. Extensive and intensive efforts, world-wide, since 1983 have not succeeded in making the high temperature

properties of Nd-Fe-B based magnets comparable with those of either the 1:5's or 2:17's. And experience has shown that at least several years would be required to develop any new permanent magnet material into a commercially available product.

There are at least several reasons why there has been little improvement in the permanent magnet properties of uncompensated 2:17-type permanent magnets since 1979. The preparation and compositional control of the multicomponent 2:17 alloys, their comminution into fine powders, magnetic alignment and compaction of the powders, sintering into dense bodies, and subsequent heat treatments are significantly more complex than the corresponding steps for 1:5-type magnets, and much more sensitive to minor compositional and processing variations. The metallurgical behavior of the complex alloy systems and the strongly interactive nature of the magnet compositions with the subsequent heat treatments required to develop superior magnetic properties were poorly understood, so that early attempts to simplify the processing and/or improve the magnetic properties of the 2:17's were unsuccessful. Finally, the promise of the Nd-Fe-B type magnets to replace the 1:5 and 2:17 types with less expensive, easier to manufacture, and significantly higher energy products very effectively diverted much of the attention of the world's magnet manufacturers and research laboratories away from the difficult problems associated with the further development of the 2:17's. Indeed, this ARO supported effort at the University of Dayton, plus a 2:17-type permanent magnet manufacturing development program by a privately-owned magnet manufacturing company involving the Principal Investigator as a consultant (see Ref. 1), appear to have been the only active programs directed to the development of 2:17-type permanent magnets in the USA during the period of the contract.

The primary objective of this research effort has been to test and refine a model proposed by the Principal Investigator to describe the metallurgical behavior of $\text{Sm}(\text{Co,Fe,Cu,Zr})_2$ alloys and the complex relationships between the compositions, heat treatments, microstructures and compositions of the phases

observed, and the corresponding magnetic properties developed by the alloys. A second objective was to develop our laboratory techniques and procedures for the processing of 2:17 type permanent magnet in order to be reasonably confident that observed variations in the magnetic and other physical properties we measured would reflect intentional changes in compositional and heat treating parameters. We did not accomplish a third objective of developing higher energy product 2:17 type permanent magnets than were known at the beginning of the research effort.

REFERENCES TO PROBLEM STATEMENT

1. Marlin H. Walmer, "A Comparison of Temperature Compensation in SmCo_5 and $\text{RE}_2(\text{TM})_{17}$ as Measured in a Permeameter, a Traveling Wave Tube, and an Inertial Device Over the Temperature Range of -60°C to 200°C ," Paper No. W3.4 at the 9th International Workshop on Rare Earth Magnets and Their Applications, Bad Soden, FRG, August 31-September 2, 1987. (Proceedings Book by: Deutsche Physikalische Gesellschaft e.V., D-5340 Bad Honnef 1, FRG). Address inquiries to Marlin H. Walmer, Electron Energy Corporation, P.O. Box 458, Landisville, PA, 17538.
2. A.E. Ray, "The Development of High Energy Product Permanent Magnets From 2:17 RE:TM Alloys," IEEE Trans. Magn., MAG-20, (1984) 1614.
3. T. Yoneyama, A. Fukuno, and T. Ojima, " $\text{Sm}_2(\text{Co,Cu,Fe,Zr})_{17}$ Magnets Having High H_c and $(BH)_{\text{max}}$," Proc. 3rd. Int. Conf. on Ferrites, Kyoto, Japan (1980). See also R.K. Mishra, G. Thomas, T. Yoneyama, A. Fukuno, and T. Ojima, "Microstructure and Properties of Step Aged Rare Earth Alloy Magnets," J. Appl. Phys. 48, (1977) 1350.

SUMMARY OF IMPORTANT RESULTS

1. While the basic concepts underlying the original model for the metallurgical behavior of the multi-component, 2:17-type $\text{Sm}(\text{Co}, \text{Fe}, \text{Cu}, \text{Zr})_2$ alloys in the development of high coercivity and high energy products remain valid, the model itself has undergone several significant revisions and has become much more detailed. (Please refer to Paper nos. 1, 2, 4, and 6 in the List of Publications and Reports).

2. A major research project was undertaken to determine the individual and interactive effects of sintering atmosphere, sintering temperature, sintering time, solid solution heat treatment temperature, and solid solution heat treatment time, on grain size, density, and saturation magnetization of 2:17 magnets of a standard composition corresponding to $\text{Sm}_2(\text{Co}_{.62}\text{Fe}_{.28}\text{Cu}_{.06}\text{Zr}_{.02}\text{Vac}_{.02})_{17}$ for the metallic component of the magnets. The results of this study are contained in J.L. Calvert's M.S. Thesis (Paper No. 5 in the List of Publications and Reports).

3. During the first two years of this project, it became clear that verification of, or alternate explanations for, significant details of the model proposed for the metallurgical behavior of the 2:17 alloys required detailed AEM and high resolution TEM analyses be conducted with state-of-the-art electron optical equipment on compositionally, metallurgically, and magnetically well-characterized magnet alloy samples. The samples were already available from our previous ARO Program (Contract No. DAAG-81-K-0120, 17973-MS). By way of a subcontract, we enlisted the support and cooperation of Prof. W.A. Soffa, Prof. J.R. Blachere, and their graduate assistant, Mrs. Bing Zhang of the Department of Materials Science and Engineering, University of Pittsburgh. Prof. Soffa and his group had recently acquired a new JEOL 200CX high resolution TEM and a JEOL 2000FX AEM. The first results of this cooperative effort were very successful

and are described in Paper No. 6. The abstracts of two additional papers (No. 7 and No. 8) have been accepted for presentation at the 9th International Rare Earth Permanent Magnet Workshop in Bad Soden (FRG) 31 Aug.-3 Sept. 1987. It is unfortunate we are unable to continue this fruitful relationship under ARO sponsorship.

LIST OF PUBLICATIONS AND REPORTS

1. A.E. Ray, "Metallurgical Behavior of $\text{Sm}(\text{Co}, \text{Fe}, \text{Cu}, \text{Zr})_2$ Alloys," J. Appl. Physics, 55 (1984) 2094-2096.
2. A.E. Ray, "The Development of High Energy Product Permanent Magnets from 2:17 RE:TM Alloys," IEEE Trans. Magn., MAG-20 (1984) 1614-1618.
3. D. Li and K.J. Strnat, "Domain Behavior in Sintered Nd-Fe-B Magnets During Field Induced and Thermal Magnetization Change," J. Appl. Phys. 57 (1985) 4143-4145.
4. A.E. Ray, "The Development of High Coercivity in 2:17 Type Rare Earth-Transition Metal Permanent Magnets," pp. 105-118 in Soft and Hard Magnetic Materials with Applications. Edited by J. A. Salsgiver, et al, Published by American Society for Metals, Metals Park, Ohio, October, 1986.
5. J.L. Calvert, "A Study of the Effects of Heat Treatment Variables on Microstructural, Physical, and Magnetic Properties of $\text{Sm}(\text{Co}, \text{Fe}, \text{Cu}, \text{Zr})_2$ Alloys," Thesis (M.S.) submitted to the Department of Materials Engineering, School of Engineering, University of Dayton, December, 1986.
6. A.E. Ray, W.A. Soffa, J.R. Blachere, and B. Zhang, "Cellular Microstructure Development in $\text{Sm}(\text{Co}, \text{Fe}, \text{Cu}, \text{Zr})_{8.35}$ Alloys," Paper No. DC-04 presented at INTERMAG '87, Tokyo, Japan, April, 1987. To be published in IEEE Trans. Magn., MAG-23 September (1987).
7. B. Zhang, J.R. Blachere, W.A. Soffa, and A.E. Ray, "Compositional and Structural Analysis of the Cellular Microstructure Development in 2:17 Type Permanent Magnet

Alloys by Analytical Electron Microscopy," (Abstract only).
Paper No. WP7.1 to be presented at the Ninth International
Workshop on Rare Earth Magnets, Bad Soden, FRG, 31 August-2
September 1987.

8. A.E. Ray, W.A. Soffa, J.R. Blachere, and B. Zhang, "A
Revised Model for the Metallurgical Behavior of 2:17 Type
Permanent Magnet Alloys," (Abstract only). Paper No.
WP7.2, *ibid.*

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1. Alden E. Ray (Principal Investigator), Professor of Materials Engineering, School of Engineering, Senior Metallurgist and Supervisor of the Metals and Ceramics Division, Research Institute.
2. Karl J. Strnat, Tait Professor of Electrical Engineering and Director of the Magnetics Laboratory, School of Engineering.
3. Herbert J. Mildrum, Adj. Professor of Electrical Engineering, School of Engineering and Research Engineer, Research Institute.
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University of Pittsburgh

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1. William A. Soffa, Professor
2. Jean R. Blachere, Associate Professor
3. Bing Zhang, Graduate Research Assistant

Jeffrey L. Calvert was awarded the degree of Master of Science in Materials Engineering while participating on this project.

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